Modulated arsenic molecular-beam epitaxial growth of $In_{0.48}Al_{0.52}As$

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A modulated As molecular-beam epitaxy method is utilized to grow high-quality $In_{0.48}Al_{0.52}As$ epilayer at a low substrate temperature, which is compatible to the optimal $In_{0.47}Ga_{0.53}As$ growth condition. The reflection high-energy electron diffraction intensity oscillations recorded during modulated As beam epitaxy shows that a persistent layer-by-layer growth mode is maintained throughout the epitaxy process. The high electron mobility, high photoluminescence (PL) intensity, and narrow PL linewidth have confirmed the low defect generation/incorporation during the modulated As beam epitaxy of the $In_{0.48}Al_{0.52}As$. The effect of interface smoothing is confirmed by the extremely narrow 77 K PL linewidth of an $In_{0.47}Ga_{0.53}As/In_{0.48}Al_{0.52}As$ quantum well stack heterostructure grown by this method.

I. INTRODUCTION

In preparing the $In_{0.47}Ga_{0.53}As/In_{0.48}Al_{0.52}As$ heterostructure by molecular-beam epitaxy (MBE), the incompatible growth conditions make it very difficult to obtain high-quality $In_{0.47}Ga_{0.53}As$ and $In_{0.48}Al_{0.52}As$ layers simultaneously. The high-quality $In_{0.48}Al_{0.52}As$ can be achieved only at a high growth temperature (>550 °C) where a smooth growth front is maintained.[1] On the other hand, most of the reported high-quality $In_{0.47}Ga_{0.53}As$ layers were grown at a medium substrate temperature (≤520 °C) with high V/III flux ratio.[2] The high substrate temperature will cause strong indium desorption during the $In_{0.47}Ga_{0.53}As$ growth,[3] while the low substrate temperature and high arsenic overpressure will cause a rough growth front during the $In_{0.48}Al_{0.52}As$ growth, owing to the low surface mobility of Al adatoms.[4] The rough growth front causes three-dimensional growth and defect generation/incorporation, and it is detrimental to both the electrical and optical properties of the semiconductor.[5,6] Although the conventional growth interruption method can provide a smooth growth front, the long surface recovery time for $In_{0.48}Al_{0.52}As$, larger than 3 min,[7] is impractical and results in a high level of impurity incorporation during the long interruption period. The surface mobility of Al adatoms can be enhanced either by increasing substrate temperature or by lowering the As overpressure.[8,9] Unfortunately, both methods are impractical when growing high-quality $In_{0.47}Ga_{0.53}As/In_{0.48}Al_{0.52}As$ heterostructures, where a relatively low substrate temperature and a high V/III flux ratio are needed during the growth of $In_{0.47}Ga_{0.53}As$. This dilemma can be solved by using a modulated As molecular-beam epitaxy method similar to that reported for the growth of the GaAs/AlGaAs heterojunction at a low substrate temperature.[10]

II. EXPERIMENT

In this study, the As overpressure is set to comply with the optimal growth condition of the $In_{0.47}Ga_{0.53}As$, but is lowered by periodically opening and closing the As shutter while keeping the Al and In shutters constantly open. During the period of which the As shutter is closed, the Al and In adatoms impinging onto the substrate surface will react with the residual As molecules inside the growth chamber. This low V/III flux ratio condition will enhance the surface mobilities of adatoms during the $In_{0.48}Al_{0.52}As$ growth and result in a smooth growth front. After an optimal duration, when the As shutter is reopened, the As overpressure is gradually increased to the originally calibrated V/III flux ratio for the high-quality $In_{0.47}Ga_{0.53}As$ growth. Therefore, when the modulating sequence is optimized, the adequate V/III flux ratio can be maintained simultaneously for both $In_{0.47}Ga_{0.53}As$ and $In_{0.48}Al_{0.52}As$ growths. In addition, unlike the conventional growth interruption method or the migration enhancement epitaxy method, the growth rate of the modulated As beam method is not affected by the periodic As shutter operation.

All samples are grown on Fe-doped InP (100) on-axis substrates. Before epilayer growth, the substrate is mounted on a molybdenum (Mo) block with indium, and thermally desorbed in the growth chamber. The desorption is completed when a (2×4) reflection high-energy electron diffraction (RHEED) pattern is observed. The growth is then carried out at a substrate temperature of ~520 °C. The substrate temperature is monitored simultaneously by an infrared pyrometer and a thermocouple in contact with the Mo block. The infrared pyrometer reading was calibrated previously with the melting temperature of an InSb wafer. The thermally cracked solid As with a dimer to tetramer ratio of >5 is used as the As source. The $As_2/(Al+In)$ flux ratio was determined before the growth with a quadrupole mass spectrometer placed at the growth position and is fixed at about 25:1 in all cases. The growth rate is about 0.7 μm/h for all samples and was calibrated previously by using RHEED intensity oscillations. For consistency, all epilayers are about 1 μm thick. The epilayers are grown with Al and In shutters open at all times, while the As shutter is periodically opened and closed.

III. RESULTS AND DISCUSSION

Figure 1(a) is the typical RHEED intensity oscillations recorded at the beginning of every $In_{0.48}Al_{0.52}As$ growth in this study. The oscillations can sustain only several monolayers and the oscillation intensity dies out very quickly. Appar-

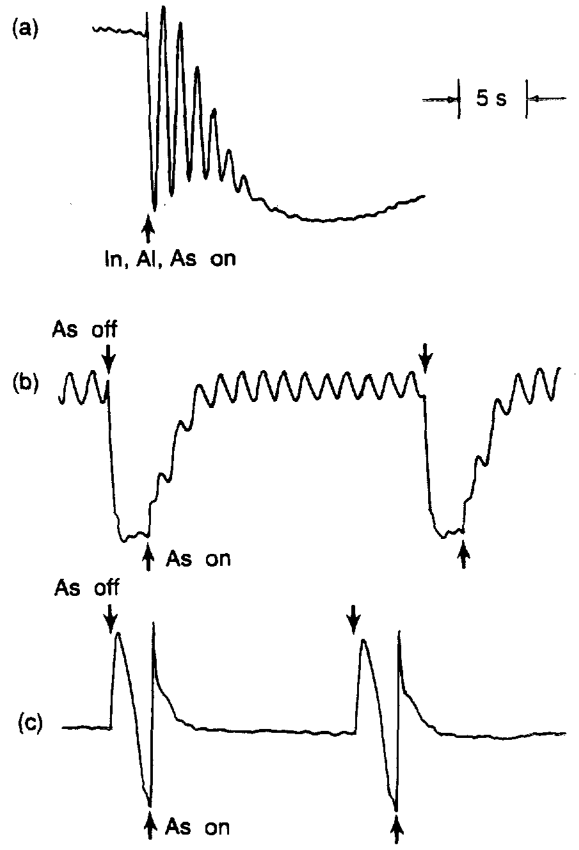

FIG. 1. The RHEED intensity oscillations recorded (a) at the beginning of the $In_{0.48}Al_{0.52}As$ growth; (b) during the modulation As beam epitaxy with a modulation sequence of 20 s As on and 3 s As off; and (c) during the modulation As beam epitaxy with a modulation sequence of 15 s As on and 3 s As off.

ently a smooth growth front can no longer be maintained and the three-dimensional (3D) growth is probably proceeding. However, when the optimal As beam modulation sequence is applied, which is 20 s As on and then 3 s As off in this study, strong RHEED intensity oscillations are maintained as shown in Fig. 1(b). This persistent RHEED intensity oscillations throughout the whole growth period indicates that a layer-by-layer growth mode has been reached at all times.[6] On the other hand, when nonoptimal modulation sequences are applied, although the oscillations can be maintained initially similar to Fig. 1(b), the RHEED oscillation intensity fades away as the growth continues. As shown in Fig. 1(c), no RHEED intensity oscillations are observed during the growth of the $In_{0.48}Al_{0.52}As$ layer using a nonoptimal As beam modulation sequence of 15 s As on and 3 s As off, indicating that 3D growth is probably in process.

Samples grown by the conventional MBE method and the modulated As epitaxy method with an optimized modulation sequence show a small lattice mismatch of less than 5×10^{-4}, as determined by the double-crystal x-ray diffraction (DCXRD) measurement. Those samples grown with nonoptimal modulation sequences show a mismatch in the 5×10^{-4} to 1×10^{-3} range. The unintentionally doped $In_{0.48}Al_{0.52}As$ layer grown by the modulated As beam method with an optimal As modulation sequence shows n-type conductivity

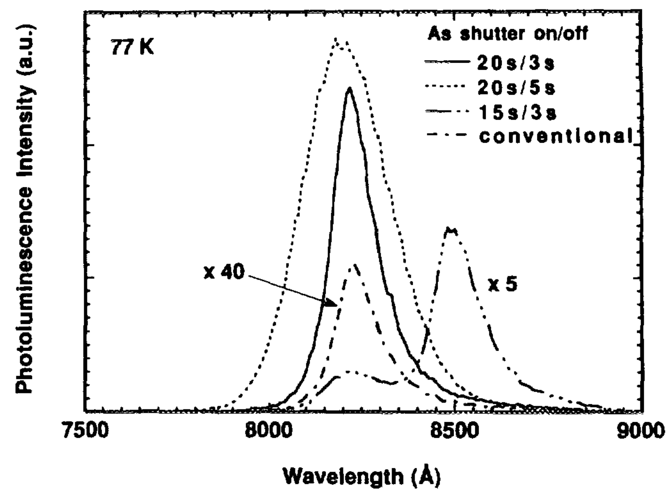

FIG. 2. PL spectra measured at 77 K in some typical $In_{0.48}Al_{0.52}As$ samples. The intensities of samples grown by the conventional MBE method and the nonoptimal modulation As beam epitaxy (15 s As on and 3 s As off) have been multiplied by a factor of 40 and 5, respectively.

with a background carrier concentration of $\sim5\times10^{15}$ cm^{-3} and electron mobilities of 1900 and 3900 cm^2/V s at room temperature and 77 K, respectively, as determined by the van der Pauw Hall measurement. These numbers are comparable to the best value reported for samples grown at high substrate temperatures (>550 °C) or with an $In_{0.47}Ga_{0.53}As$ buffer layer.[11,12] However, samples grown at ~520 °C by the conventional MBE method or by nonoptimal As modulation sequences show either semi-insulating properties or exhibit low electron mobilities. With a laser excitation source tuned to 5145 Å, 77 K photoluminescence (PL) has been measured from these samples. As shown in Fig. 2, the samples grown with the optimal condition (20 s As on and 3 s As off) show a narrow linewidth with a full width at half-maximum (FWHM) of about 20 meV, similar to the samples grown by the conventional method. Its intensity is about 100 times higher than that grown by the conventional method. When grown with the nonoptimal modulation sequences, the samples show multiple PL peaks and much larger linewidths.

To further investigate the sharpness of the interfaces prepared by the modulated As beam method, an $In_{0.47}Ga_{0.53}As/In_{0.48}Al_{0.52}As$ quantum well stack structure with different well widths is grown. The structure consists of a 0.5 μm thick $In_{0.48}Al_{0.52}As$ buffer layer, a 0.2 μm $In_{0.48}Al_{0.52}As$ cap layer, and several $In_{0.47}Ga_{0.53}As$ quantum wells separated by 500 Å thick $In_{0.48}Al_{0.52}As$ barrier layers. Figure 3 is the PL spectra of such a quantum well stack heterostructure measured at 77 K. The FWHMs are 17, 9, 5.7, 10, and 12 meV for the 15, 40, 60, 100, and 140 Å quantum wells, respectively. These narrow FWHMs are comparable to the best reported low-temperature (<15 K) values of samples grown either with growth interruptions or at a high substrate temperature.[13,14]

IV. CONCLUSIONS

A modulated As beam method to grow high-quality $In_{0.48}Al_{0.52}As$ under a growth condition compatible to the

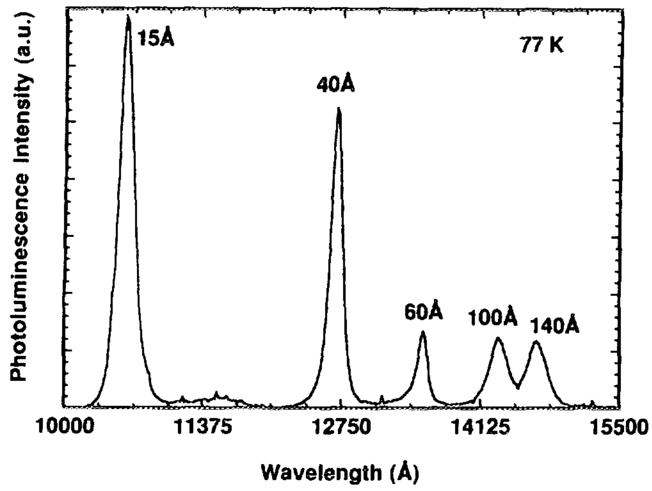

FIG. 3. PL spectra measured at 77 K on an $In_{0.47}Ga_{0.53}As/In_{0.48}Al_{0.52}As$ quantum well stack heterostructure.

optimal $In_{0.47}Ga_{0.53}As$ growth condition has been developed. By optimally modulating the As shutter, a smooth growth front can be maintained during the growth of $In_{0.48}Al_{0.52}As$ without using an $In_{0.47}Ga_{0.53}As$ buffer layer or growth interruptions. In addition, the growth rate is not affected by the As shutter modulation. The persistent layer-by-layer growth mode affirmed by the RHEED intensity oscillations obviously has decreased the defect formation/incorporation as reflected by the measured high electron mobility, high PL intensity, and narrow PL linewidth. The improvement of the surface smoothness is further confirmed by the 77 K PL spectrum measured in an $In_{0.47}Ga_{0.53}As/In_{0.48}Al_{0.52}As$ quantum well stack heterostructure grown by the modulated As beam method. The extremely narrow linewidth is comparable to the best low-temperature PL results reported for this system.

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